RECOMMENDATIONS FOR IMPROVING THE INTERIM RADIONUCLIDE SOIL ACTION LEVELS FOR THE ROCKY FLATS CLEANUP AGREEMENT

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DOE Environmental Management Science Program (EMSP) Project

Improved Radiation Dosimetry/Risk Estimates to Facilitate Environmental Management of Plutonium-Contaminated Sites (A Second Year Project)

INTRODUCTION

Our Institute is conducting research for the Department of Energy's (DOE) Environmental Management Science Program (EMSP) to develop "improved dosimetry/risk estimates to facilitate environmental management of plutonium-contaminated sites." Our initial focus was on evaluating radioactivity intake for the stochastic, nuclear-worker exposure paradigm where variability in PuO2 intake associated with particle polydispersity (size variability) was found to be very important. The importance arose because of the very high specific activity of some pure PuO₂ particles (e.g., ²³⁸PuO₂, ²³⁹PuO₂, ²⁴⁰PuO₂) and because particle radioactivity increases roughly in proportion to the cube of the particle size. The findings relate more to workers involved in decommissioning/decontamination activities at Pu-contaminated sites such as the Rocky Flats Environmental Technology Site near Denver, Colorado. More recently our work has focused on inhaled PuO2-contaminated, resuspended dust. The specific activity of the contaminated dust, however, is orders of magnitude lower than for pure PuO2. Our interest in inhaled contaminated, resuspended dust relates to soil at Rocky Flats. A key issue is what level of cleanup is needed at Rocky Flats so that the public would not be faced with unacceptable risks for harm from inhaled Pu. A related issue is how important is variability in the intake of PuO₂-contaminated soil that is associated with polydispersity of dust particle size. Our poster presentation focuses on characterizing variability in Pu intake due to polydispersity or resuspended dust particles contaminated with PuO2. We report on implications of our research results for cleanup of the Rocky Flats site.

ISSUES

A dose standard has been proposed to control the amount of radionuclides (plutonium and other radionuclides) that may legally remain in the soil at Rocky Flats after cleanup. An action level (Tier I) triggers remedial action (e.g., soil removal) when soil radioactivity exceeds a specified level.

Proposed dose standard for Rocky Flats: Over the next 1000 years, radionuclides remaining in the surface soil may expose an *office worker* in the site's industrial zone to an annual dose of no more than 15 millirem in excess of natural background. Over the same period, a future resident farming on the site (in buffer zone) may be exposed to an annual dose of no more than 85 mrem above background.

More stringent Superfund regulations that now govern cleanup of the site would be replaced by the proposed dose standard. Adopting the standard would set a precedent.

Some claim that adopting the proposed standard will result in at least three times as many cancers as is permitted under the Superfund law that now regulates cleanup at Rocky Flats.

Major uncertainties relate to projecting health risk from environmental plutonium. The uncertainties include lack of knowledge about the shape of the risk vs. dose curve and about the variability in intake of airborne PuO_2 -contaminated soil by different individuals when inhaled over many years.

ROCKY FLATS

The Rocky Flats Environmental Technology Site is owned by the U.S. Department of Energy. The site manufactured components for nuclear weapons for the nation's defense until 1992. Its current mission is environmental cleanup, waste management, and decommissioning. The industrial complex of more than 100 buildings is located in the center of about nine square miles of undeveloped land, 16 miles northwest of downtown Denver. About 6,000 people work at the site. Rocky Flats is the repository of about 14 tons of plutonium (Pu), a radioactive metal used in nuclear bombs. Plutonium oxide (PuO₂) is present in the environment and is being cleaned up. Plutonium isotopes of concern in soil include Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242.

Workers involved in decommissioning/decontamination activities at Rocky Flats sometimes must work in 100% lethal air concentrations of PuO₂. The workers are well protected, although the specific respirator type to use for a given operation is presently being researched. Some respirators are more protective than others, and cost varies for the different types.

PLUTONIUM FACTOIDS

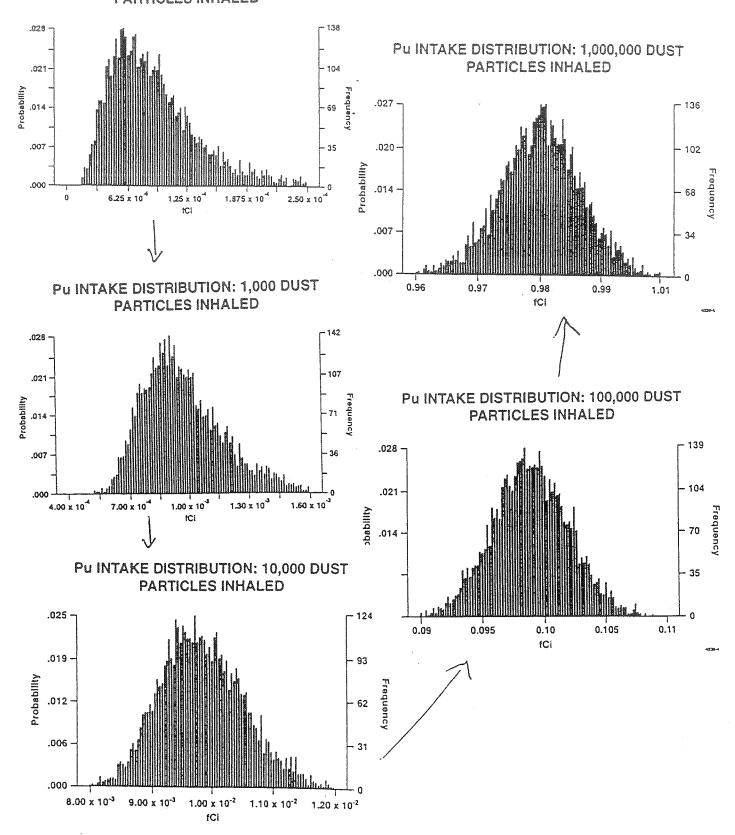
- Plutonium has occurred naturally, but except for trace quantities is not found in the earth's crust.
- Plutonium is produced in nuclear reactors and is used in weapons.
- The main isotopes of plutonium are Pu-238 (half-life \cong 88 y), Pu-239 (half-life \cong 24,000 y), Pu-240 (half-life \cong 6500 y), Pu-241 (half-life \cong 14 y), and Pu-242 (half-life \cong 376,000 y).
- Pu-238, Pu-239, Pu-240, and Pu-242 emit alpha particles, whereas Pu-241 emits beta particles.
- There are several tons of plutonium in our biosphere, a legacy of atmospheric weapons testing during the 1950s and 1960s.
- Environmental plutonium is most often in the oxide (PuO₂) form.
- The major mode of intake of plutonium by humans is via inhalation.

- Russian workers at the Mayak plutonium production facility inhaled small to large amounts of PuO₂ from the late 1940s through mid-1950s.
- Nuclear workers at the Puducah [Kentucky] Gaseous Diffusion Plant inhaled unknowingly small amounts of PuO₂.
- Potential health effects from inhaled moderate amounts of PuO₂ include lung, liver, and bone cancer as demonstrated by Mayak workers.
- It is uncertain whether a very small amount of inhaled PuO₂ can cause cancer.
- A potential serious, threshold-dose type, health effect from inhaled large amounts of PuO₂ is plutonium pneumosclerosis, which was reported in Mayak workers.

APPROACH TO EVALUATING VARIABILITY IN PuO₂ INTAKE IN INHALED ROCKY FLATS SOIL

The specific activity of the PuO_2 -contaminated dust (soil) at Rocky Flats is orders of magnitude lower than for pure PuO_2 so that large numbers of dust particles must be inhaled to lead to significant radiation exposure. Starting from single dust particle intake, conditional, single-particle and multiple-particle intake distributions were generated for generic PuO_2 inhaled in resuspended Rocky Flats soil. Intake distributions were generated for adult males who inhaled and deposited in the respiratory tract 100, 200, 500, 1,000, 2,000, 10,000, 100,000, 1,000,000, or 5,000,000 PuO_2 -contaminated dust particles. Results were developed for soil specific activity of 1 Bq/g (27 pCi/g), soil density of 2 g/cm³, and for a polydisperse size distribution (truncated lognormal: AMAD = 1 μ m; σ_g = 2.5; maximum aerodynamic diameter = 26 μ m). This allows easy scaling to other soil specific activities. We expect our results for inhaling soil contaminated with PuO_2 to apply for a first approximation to adult females. Deposition efficiencies were evaluated based on ICRP 66. Monte Carlo evaluations of intake distributions were carried out using Crystal Ball software.

Pu INTAKE DISTRIBUTION: 100 DUST PARTICLES INHALED



Intake-Distribution-Related Statistics for PuO₂ Inhaled in Rocky Fiats Soil by Adult Males (Soil specific activity = 27 pCi/g)

Number of contaminated dust particles inhaled	Mean intake in picocuries	Skewness	Kurtosis	Coefficient variation	Max/mean
100	9.67x10 ⁻⁶	2.63	13.2	0.65	6.92
200	1.93x10 ⁻⁷	2.20	10.6	0.47	4.13
500	4.91×10 ⁻⁷	1.62	7.20	0.33	3.44
1,000	9.86×10 ⁻⁷	1.16	3.45	0.23	2.34
2,000	1.97×10°	0.76	3.86	0.16	1.71
10,000	9.83×10 ⁻⁵	0.35	3.16	0.073	1.31
100,000	9.83x10 ⁻⁵	0.11	2.97	0.023	1.09
1,000,000	9.83x10 ⁻⁴	0.06	2.88	0.0075	1.03
5,000,000	4.91×10 ⁻³	0.03	3.07	0.0033	1.01

1 picocurie = 1x10⁻¹² curies.

 Skewness values greater than zero imply a tail to the right. The larger the skewness, the longer the tail of the distribution.

Kurtosis values indicate the steepness of the peak. Steepness of the peak of the distribution increases as the kurtosis increases.

Mean Intake of PuO₂ in Resuspended Rocky Flats Soil Inhaled by Adult Males as Function of the Number of Dust Particles Inhaled and Average Soil Specific Activity

Number of contaminated dust particles	Radioactivity intake (pCi) by soil average specific activity				
inhaled	27 (pCi/g)	270 (pCi/g)	2,700 (pCi/g)	27,000 (pCi/g)	270,000 (pCi/g)
100	9.67×10 ⁻⁸	9.67x10 ⁻⁷	9.67x10 ⁻⁶	9.67x10 ⁻⁵	9.67x10 ⁻⁴
200	1.93×10 ⁻⁷	1.93x10 ⁻⁶	1.93x10 ⁻⁵	1.93x10 ⁻⁴	1.93x10 ⁻³
500	4.91x10 ⁻⁷	4.91x10 ⁻⁶	4.91x10 ⁻⁵	4.91x10 ⁻⁴	4.91x10 ⁻³
1,000	9.86x10 ⁻⁷	9.86x10 ⁻⁶	9.86x10 ⁻⁵	9.86x10 ⁻⁴	9.86x10 ⁻³
2,000	1.97×10 ⁻⁵	1.97x10 ⁻⁵	1.97x10 ⁻⁴	1.97x10 ⁻³	1.97x10 ⁻²
10,000	9.83x10 ⁻⁶	9.83x10 ⁻⁵	9.83x10 ⁻⁴	9.83x10 ⁻³	9.83x10 ⁻²
100,000	9.83x10 ⁻⁵	9.83x10 ⁻⁴	9.83x10 ⁻³	9.83x10 ⁻²	0.983
1,000,000	9.83x10 ⁻⁴	9.83x10 ⁻³	9.83x10 ⁻²	0.983	9.83
10,000,000	9.83x10 ⁻³	9.83x10 ⁻²	0.983	9.83	98.3
100,000,000	9.83x10 ⁻²	0.983	9.83	98.3	983
1,000,000,000	0.983	9.83	98.3	983	9,830

• 1 pCi = 1x10⁻¹² curies.

Single Radionuclide Soil Action Levels for Rocky Flats*

	Hypothetical office worker exposure scenario (Tier I): Limit 15 mrem/y	Hypothetical residential exposure scenario (Tier I): Limit 85 mrem/y	Hypothetical residential exposure scenario (Tier II): Limit 15 mrem/y
Radionuclide	pCi/g	pCi/g	pCi/g
Plutomium-238	1164	1529	270
Plutonium-239	1088	1429	252
Plutonium-240	1089	1432	253
Plutonium-241	7801	19830	3499
Plutonium-242	1145	1506	266

Tier I: remedial action and/or management required if exceeded given the presence of institutional controls.

Tier II: no remedial action and/or institutional control required if not exceeded *Information from "Action Levels for Radionuclides in Soils for the Rocky Flats Cleanup Agreement: Final" 1996.

Inhalation Intake to Dose Conversion Coefficients in mrem/pCi*

Plutonium isotope	Bone			Red
	surface	Liver	Lung	marrow
Pu-236	0.837	0.165	0.692	0.0670
Pu-238	2.69	0.507	1.19	0.215
Pu-239	3.04	0.559	1.20	0.243
Pu-240	3.04	0.559	1.20	0.243
Pu-241	0.0659	0.0111	0.0118	0.0530
Pu-242	0.289	0.530	1.14	0.231
Pu-244	2.85	0.526	1.12	0.229

^{*}For the slow, "S", absorption class. Based on information from Federal Guidance Report 11; available on the web at:

http://www.ornl.gov/~wlj/fgr11tab.htm. Bqs were converted to pCi using 1Bq = 27 pCi.

file: RFE. 3.4. 2 Soil action Levels Reports

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MEMORANDUM

9/2/99

FROM:

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TO:

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Enclosed are a recent publication (Radiat. Prot. Dosim 83(3), 221-232, 1999) and a poster presentation that I thought you and/or your colleagues may find of interest that relate to worker and public exposures to Rocky Flats plutonium.

The publication is entitled "Variability in PuO₂ intake by inhalation: Implications for worker protection at the US Department of Energy." You may recall having received a prepublication copy in April of this year with a slightly different title. The journal (published in the UK) changed the title just before publication without my knowledge to clarify that the paper relates to the US DOE.

The poster presentation is entitled "Recommendations for improving the interim radionuclide soil action levels for the Rocky Flats Cleanup Agreement" and was given in New Orleans on 8/22/99 and again on 8/23/99 at the 218th National Meeting of the American Chemical Society. We plan to make the poster available on the web.

Our research findings so far support those stated in the Colorado Department of Public Health and Environment report entitled "Historical Public Exposures Studies on Rocky

Flats" as cited in Weapons Complex Monitor Volume 10, No. 32. More specifically, our research results support the following views:

- People who were in the path of airborne plutonium releases from a fire on Sept. 11-12, 1957, were likely subjected to the highest risk of all the Rocky Flats plant releases.
- A person's location, lifestyle and period of exposure likely have a greater effect on health risks than age or gender.

Based on our research, we would expect that large variability in plutonium intake (via inhaled, resuspended, contaminated dust) could arise because of differences in lifestyle and variability in the length of exposure. However, variability in intake due to differing dust particle sizes (polydispersity) was found to be unimportant for public exposure to contaminated dust over long periods. Very large numbers of PuO_2 -contaminated dust particles were calculated to be needed in order to have significant intake of radioactivity (unlike for pure PuO_2). With very large numbers of contaminated dust particles inhaled, variability in intake due to polydispersity of size is calculated to be unimportant.

Regarding public exposure arising from the 1957 fire, or research results indicated that large variability in the PuO_2 intake (due to polydispersity of particle size) should have occurred as relatively small numbers of pure PuO_2 particles would likely have been inhaled. This is what we refer to as the stochastic-exposure paradigm in our paper.

Rocky Flats/ROC

9/2/1999

Rocky Flats Environmental - Soil Issues - Soil Action Levels - Reports - RECENT PUBLICATION-POSTER PRESENTATION PUBLIC EXPOSURES PLUTONIUM

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